

# Biogeochemistry of Himalayan rivers as an agent of climate change

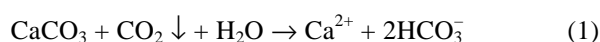
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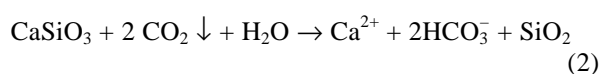
**Uplift of the Himalaya, intensification of the Asian monsoon and increased erosion from the Himalayan orogen are all intimately coupled and reflected in the transport of weathering products to the ocean by the large rivers, namely the Indus, Ganga, Brahmaputra, Mekong, Chang Jiang and Irrawady<sup>1-6</sup>. In recent years, several studies have focused on the coupling between continental weathering rates and changes in atmospheric CO<sub>2</sub> budget. Because the atmospheric CO<sub>2</sub> reservoir is relatively small and CO<sub>2</sub> gets converted to bicarbonate anion (HCO<sub>3</sub><sup>-</sup>) and is balanced by dissolved cations from the silicate/carbonate rocks; it is likely that this reservoir is sensitive to changes induced by chemical weathering rates. In such a scenario, increased chemical weathering and associated CO<sub>2</sub> consumption rates in the Himalayan rivers could alter the atmospheric CO<sub>2</sub> levels and hence the global climate. Both oceanic and terrestrial evidence points to a marked, progressive period of intense cooling of global climate over the geologic past. Existing models of the global CO<sub>2</sub> cycle do not include detailed consideration of such tectonically-driven enhanced chemical weathering. The purpose of this paper is to evaluate the present-day CO<sub>2</sub> consumption rates by chemical weathering processes in the Himalaya, based on newer data<sup>6</sup>, and their implications to long-term climate change.**

THE long-term ( $T > 10^5$  yrs) variations in the earth's atmospheric CO<sub>2</sub> levels are driven by a variety of geologic processes. CO<sub>2</sub> is added to the atmospheric reservoir by global volcanic degassing, the main source term; and the two major processes representing as sink: the uptake of CO<sub>2</sub> during rock weathering and that during photosynthesis and transformation to organic matter. The CO<sub>2</sub> removal via organic matter formation and rock weathering (mainly carbonate and silicate weathering) is eventually converted into dissolved organic carbon (DOC); particulate organic carbon (POC) and dissolved inorganic carbon (DIC as HCO<sub>3</sub><sup>-</sup> ion) pools of the rivers; which are subsequently exported to the ocean. The generalized reactions for CO<sub>2</sub> consumption by chemical rock weathering and photosynthesis are schematically described as:

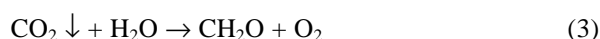
Carbonate weathering:



Silicate weathering:

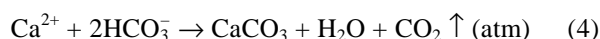


Photosynthesis:

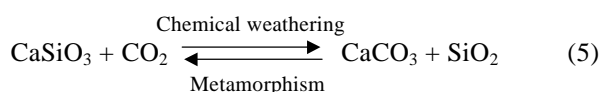


It is important to bear in mind from the above reactions, that atmospheric/soil CO<sub>2</sub> consumed by rock weathering is considered to be equivalent to the entire HCO<sub>3</sub><sup>-</sup> flux from rivers draining the silicate rocks (eq. (2)) and to half of the HCO<sub>3</sub><sup>-</sup> flux from rivers draining carbonate rocks (eq. (1)). All carbon fixed in the organic matter and transported to the rivers is of atmospheric origin (eq. (3)).

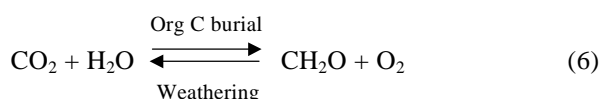
On a million-year time scale, continental weathering of carbonate minerals does not affect atmospheric CO<sub>2</sub>, as it is returned to the atmosphere during precipitation of calcite/aragonite/dolomite minerals in the oceans<sup>7</sup>, eq. (4):



By combining the reactions in eq. (2) and eq. (4), the rock weathering reactions are simplified as:



In addition to the carbonate-silicate cycle described by weathering reaction in eq. (5), the organic carbon subcycle on long-term (as described schematically by eq. (6)) has a significant impact on the cycling of atmospheric CO<sub>2</sub>.



The CO<sub>2</sub> fluxes via volcanic outgassing and silicate weathering (eq. (2)), along with organic C burial and weathering (eq. (6)) are relatively large in comparison to the total C storage at the earth's surface. It is estimated

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that the global CO<sub>2</sub> outgassing flux is approximately  $8 \times 10^{18}$  moles Myr<sup>-1</sup>, while the mass of C in the atmosphere is about  $0.06 \times 10^{18}$  moles<sup>7,8</sup>. It is thus required that the rates of CO<sub>2</sub> release via global degassing plus organic matter weathering must be closely balanced by the CO<sub>2</sub> uptake via silicate weathering and organic C burial and that there must be some stabilizing feedback for maintaining atmospheric CO<sub>2</sub> within reasonable limits<sup>9</sup>. For constant degassing and organic C burial/weathering, the tectonic uplift-induced accelerated silicate weathering rate would have had a profound effect on past atmospheric CO<sub>2</sub> levels. In fact, most of the recent history of the earth is dominated by a glacial climate, with relatively short punctuations of inter-glacial phases.

Several studies have addressed this issue and have suggested a close coupling between the level of CO<sub>2</sub> in the atmosphere and global climate over at least the last 250,000 yrs<sup>10-12</sup>, although it remains uncertain how sensitive climate is to variations in atmospheric CO<sub>2</sub>. In this context, it is well accepted that changing the atmospheric CO<sub>2</sub> consumption by continental weathering can have an influence on the earth's climate. It has been stated<sup>8</sup> that higher global temperatures and greater rainfall on the continents (caused by greater CO<sub>2</sub> degassing) should bring about enhanced CO<sub>2</sub> consumption via accelerated silicate weathering. Conversely, enhanced silicate weathering by uplifted mountains would lower the atmospheric CO<sub>2</sub> level, which in turn would lower CO<sub>2</sub> consumption rates by chemical weathering processes thereby counter-balancing the effect of the uplift and erosion of mountain ranges<sup>8</sup>. Raymo *et al.*<sup>13</sup> and Raymo<sup>14</sup> have suggested that the long-term removal rate of CO<sub>2</sub> from the atmosphere by chemical weathering is a function of relief, rather than other factors and that atmospheric temperature exerts only a weak control on global chemical erosion rates. However, weathering may have an influence on climate on a relatively short-time scale. In particular, continental erosion has been discussed as a potential candidate for being involved in the glacial/inter-glacial climate cycles during Quaternary times. Munhoven and Francois<sup>15</sup> showed that an increase in silicate weathering by a factor of 2 to 3.5 during the LGM could result in variations of atmospheric CO<sub>2</sub> that are close to those documented from the ice-core records.

### River transport data

One way to study the rate of rock weathering is to evaluate the chemical composition of river water. For Ca–Mg silicates, the riverine flux of dissolved HCO<sub>3</sub><sup>-</sup> is a direct measure of the uptake of atmospheric CO<sub>2</sub> owing to weathering. Such an approach is important to estimate the present-day inorganic and organic denudations of the continents and their associated CO<sub>2</sub> consumption. The mountainous regions are dominated by rapid mechanical

erosion which, in turn, increases the surface area of fresh minerals available for chemical attack. The chemical breakdown of detrital material in mountainous regions is enhanced by the abundance of easily weathered sedimentary silicates and the orographic rainfall on mountain slopes which flush away mechanical and chemical erosion products, thereby constantly exposing fresh minerals. These effects are most marked in the Himalaya, under the influence of greater rainfall associated with the Asian monsoon. Data from the eight largest rivers draining the Himalayan–Tibetan region (Ganga, Brahmaputra, Chang Jiang, Xijiang, Irrawady, Indus, Mekong, Huanghe) show that almost 25% of the total dissolved load reaching the ocean today comes from a watershed area that represents nearly 5% of the earth's land surface. Thus, a disproportionate fraction of the earth's chemical weathering occurs in this small region in Asia. It is likely that the late Cenozoic uplift of the Tibetan plateau would have resulted in regionally, and hence globally, higher chemical erosion rates, causing a drawdown of atmospheric CO<sub>2</sub> and global cooling<sup>16</sup>.

The enhanced continental weathering rates have been well documented based on contemporary material fluxes and those based on proxy records from ocean sediments<sup>1,14</sup>. These studies have concluded that the pronounced increase in <sup>87</sup>Sr/<sup>86</sup>Sr ratio of sea water during the past 40 Myr, as recorded in marine carbonates, is attributable to erosion in low-latitude mountainous regions such as Himalaya. Richter *et al.*<sup>17</sup> suggested that the Sr flux to the ocean has increased by 40% over this interval, with a concomitant increase in delivery of radiogenic Sr via rivers. The recent studies of the Ganga–Brahmaputra headwaters suggest that the high <sup>87</sup>Sr/<sup>86</sup>Sr ratios in these rivers are dominated by intense chemical weathering of granites and gneisses (silicates)<sup>3,18</sup>. It further concurs with the suggestion<sup>5</sup> that chemical erosion in the Himalaya has been responsible for a significant fraction of the observed increase in oceanic <sup>87</sup>Sr/<sup>86</sup>Sr ratio. Given the association between silicate weathering and relatively high <sup>87</sup>Sr/<sup>86</sup>Sr ratio in the rivers of the Himalayan–Tibetan region, it is reasonable to invoke tectonics as the main control on weathering intensity; thus increasing the erosional drawdown of atmospheric CO<sub>2</sub> leading to climate change.

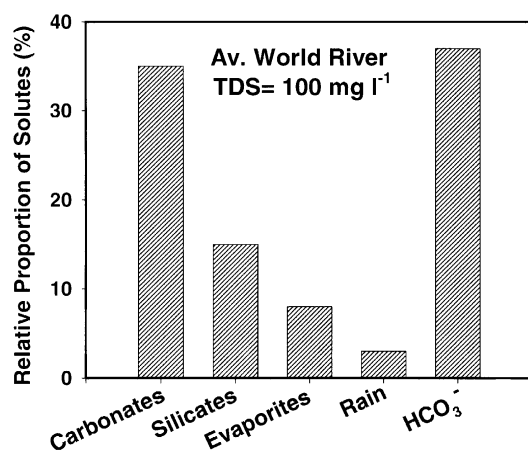
### Rock weathering and constituent fluxes

The contribution from rock weathering (silicates, carbonates and evaporite rocks), rain and atmospheric inputs dominates the dissolved load of rivers. On a global scale, relative proportions of solutes from these sources are shown in Figure 1. Solute derived from carbonate weathering and those constituting HCO<sub>3</sub><sup>-</sup> ions (derived from atmospheric CO<sub>2</sub> consumed by carbonate and silicate weathering) account for almost 70% of the dissolved load, with equal contribution from the two sources. In compa-

erison, silicate weathering constitutes no more than 15% to the dissolved composition of river water; whereas 8% of solutes is derived from evaporites and 3% from rain water (Figure 1). Based on the recent compilation of the data from 60 largest rivers, Gaillardet *et al.*<sup>6</sup> have summarized that  $300 \times 10^6$ ,  $640 \times 10^6$  and  $140 \times 10^6$  t yr<sup>-1</sup> of dissolved salts are derived from weathering of silicates, carbonates and evaporites, respectively; and that the total flux of dissolved material from continental chemical weathering is  $1080 \times 10^6$  t yr<sup>-1</sup>. Assuming that nearly 51% of the surface run-off is represented by these 60 largest rivers, Gaillardet *et al.*<sup>6</sup> have further estimated that about  $2130 \times 10^6$  t yr<sup>-1</sup> of dissolved material is derived from chemical weathering, of which about 26% ( $550 \times 10^6$  t yr<sup>-1</sup>) is contributed by silicate weathering. The component weathering fluxes for the large rivers of the Himalayan–Tibetan region have been evaluated and are compared with those of the Amazon in Figure 2. The silicate weathering fluxes for Amazon (TDS =  $80 \times 10^6$  t yr<sup>-1</sup>) and Irrawady (TDS =  $30 \times 10^6$  t yr<sup>-1</sup>) exceed the relative contribution from weathering of carbonate rocks. By far, carbonate weathering dominates the solute fluxes in the river basins of Chiang Jiang, Ganga, Brahmaputra, Indus and Mekong. It is noteworthy that carbonate weathering fluxes (TDS =  $100 \times 10^6$  t yr<sup>-1</sup>) are highest for Chiang Jiang (Figure 2). The solutes derived from evaporite weathering are not relatively significant in the Himalayan orogen.

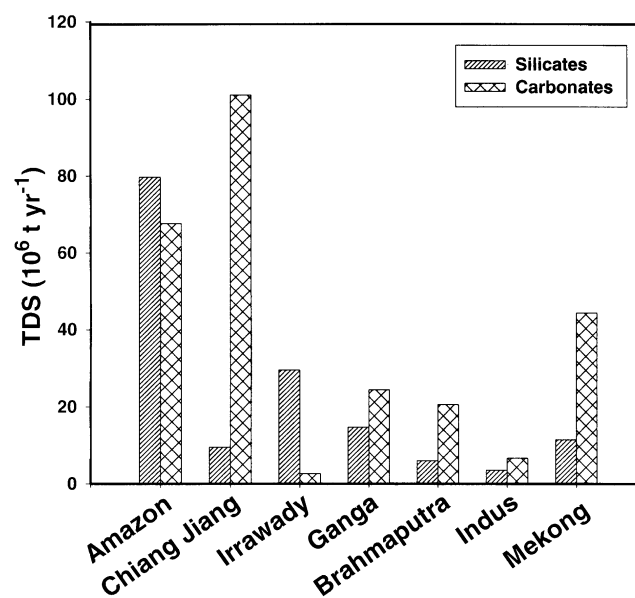
### Present-day atmospheric CO<sub>2</sub> consumption rates

The flux of atmospheric/soil CO<sub>2</sub> consumed by chemical weathering of the continents can be estimated from river fluxes of bicarbonate ion. The present-day average flux of atmospheric/soil CO<sub>2</sub> consumed by global chemical



**Figure 1.** Nearly 72% of the total dissolved salts (TDS) in the world average river water is composed of HCO<sub>3</sub><sup>-</sup> ion and those derived from carbonate weathering. Weathering of silicates and evaporites account for 15% and 8% of the solutes, respectively; whereas contribution by rain water solutes could be no more than 5%.

erosion and exported to the oceans as bicarbonate ion is estimated to be 0.28–0.30 Gt C yr<sup>-1</sup> (refs 1, 19, 20). Based on the recent compilation of data for 60 largest rivers, this paper highlights the role of Himalayan–Tibetan rivers in the global context of atmospheric CO<sub>2</sub> consumption by chemical rock weathering. In Figure 3, CO<sub>2</sub> consumption rates for carbonate and silicate weathering in the drainage basins of Chiang Jiang, Irrawady, Ganga, Brahmaputra, Indus and Mekong are compared with those for Amazon. The amount of CO<sub>2</sub> consumed by silicate weathering for the river drainage basins lying in the Himalayan–Tibetan region, ranges from (54 to 832) × 10<sup>9</sup> mol yr<sup>-1</sup>; with enhanced consumption rates for the Irrawady (832 × 10<sup>9</sup> mol yr<sup>-1</sup>) and that for Ganga (471 × 10<sup>9</sup> mol yr<sup>-1</sup>). The consumption of CO<sub>2</sub> by carbonate weathering is relatively higher for the individual rivers (except for Ganga and Irrawady) and ranges from (24 to 997) × 10<sup>9</sup> mol yr<sup>-1</sup> (Figure 3); with highest values occurring in the Chiang Jiang. These results also point out that CO<sub>2</sub> consumption rates resulting from silicate and carbonate weathering in the Himalaya are comparable in magnitude, each component contributing CO<sub>2</sub> flux of ~ 2 × 10<sup>12</sup> mol yr<sup>-1</sup>. On a global scale, the CO<sub>2</sub> consumption for silicate rock weathering is 8.7 × 10<sup>12</sup> mol yr<sup>-1</sup> (0.104 Gt C yr<sup>-1</sup>) and 12.3 × 10<sup>12</sup> mol yr<sup>-1</sup> (0.148 Gt C yr<sup>-1</sup>) for carbonate weathering<sup>6</sup>. In addition, the recent estimates show that young volcanic rocks consume 3 × 10<sup>12</sup> mol C yr<sup>-1</sup>. Thus, the present-day CO<sub>2</sub> flux consumed by chemical weathering of silicate rocks is



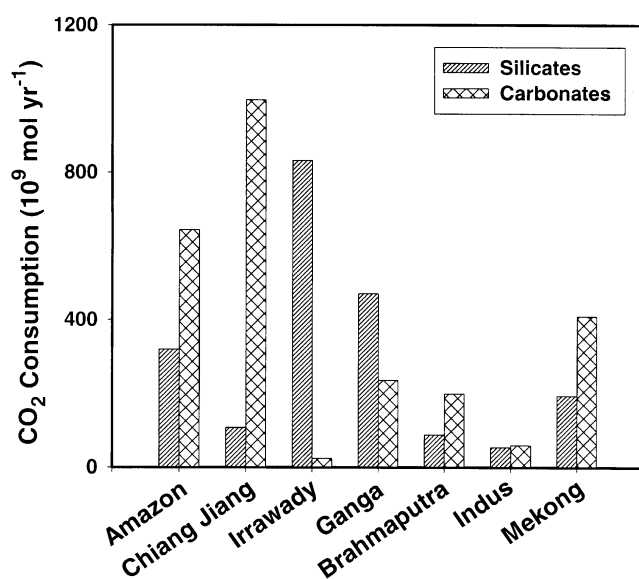
**Figure 2.** Weathering fluxes derived from silicates and carbonates for the large rivers of the Himalaya compared with those of the Amazon. Relative contribution from silicate weathering is dominant for the Amazon and Irrawady. Solute fluxes from the carbonate weathering are, in general, pronounced for most of the Himalayan rivers. For comparison, total solute fluxes from silicates and carbonates in global rivers are  $550 \times 10^6$  and  $1290 \times 10^6$  t yr<sup>-1</sup>, respectively (data from Gaillardet *et al.*<sup>6</sup>).

$11.7 \times 10^{12} \text{ mol yr}^{-1}$  ( $0.140 \text{ Gt C yr}^{-1}$ )<sup>6</sup>; of which nearly one-sixth is consumed by weathering processes in the Himalaya.

The specific  $\text{CO}_2$  consumption rates for the Himalayan-Tibetan region range from  $(146 \text{ to } 2250) \times 10^3 \text{ mol km}^{-2} \text{ yr}^{-1}$ ; with highest consumption rate for the Irrawady and lowest for the Indus (Figure 4). The specific rates for individual rivers (Figure 4) are significantly higher than those for Amazon ( $\sim 160 \times 10^3 \text{ mol km}^{-2} \text{ yr}^{-1}$ ) and for the average world rivers. Overall, the specific  $\text{CO}_2$  consumption rates (Figure 5) associated with the weathering of both silicate and carbonate rocks in the Himalayan-Tibetan rivers ( $\sim 700 \times 10^3 \text{ mol km}^{-2} \text{ yr}^{-1}$ ) are nearly three times higher than the world average river ( $\sim 250 \times 10^3 \text{ mol km}^{-2} \text{ yr}^{-1}$ ) and four to five times higher than those in the Amazon basin. Many factors such as extreme relief and monsoon rainfall result in higher weathering rates and associated  $\text{CO}_2$  consumption in the Himalaya.

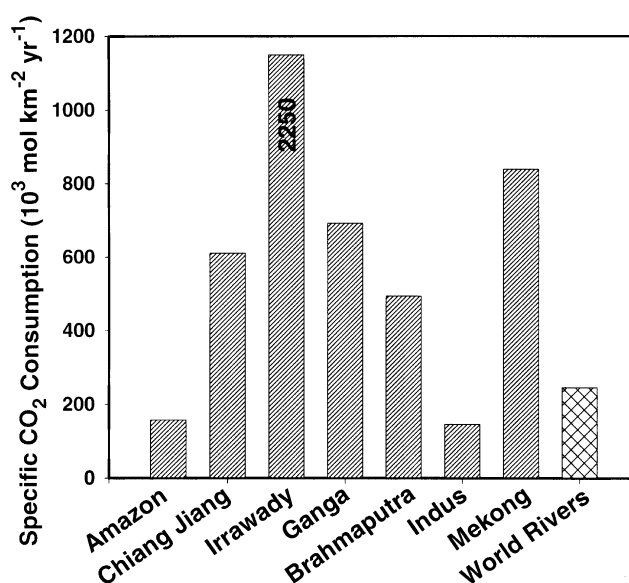
### Organic carbon burial records

The fluxes of  $\text{CO}_2$  implied by these estimates require a feedback mechanism to the atmospheric  $\text{CO}_2$  reservoir, in a scenario wherein intense erosional drawdown of  $\text{CO}_2$  could occur, as described above. The increase in silicate weathering rates could be sustained on a million-year time scale with tectonic as the main control, without an increase in  $\text{CO}_2$  mantle degassing rates. The changes in long-term burial rate of organic carbon has been suggested as the most important process causing a net addition of  $\text{CO}_2$  to the atmosphere (processes of oxidation and

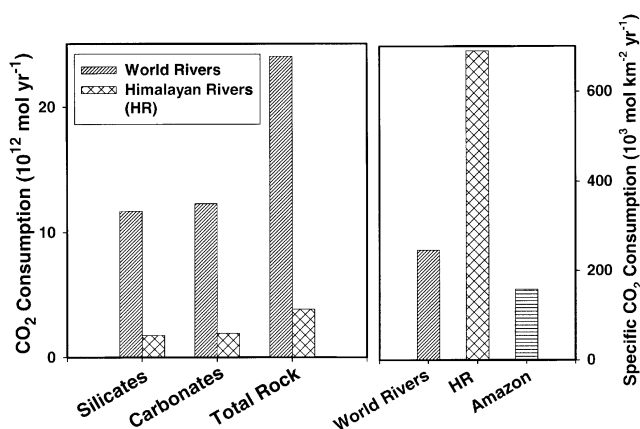


**Figure 3.**  $\text{CO}_2$  consumption rates for the Himalayan rivers compared with those of the Amazon. Enhanced  $\text{CO}_2$  consumption due to silicate weathering in the Irrawady and Ganga is noteworthy. On a global scale,  $\text{CO}_2$  consumption rates<sup>6</sup> associated with the silicate and carbonate weathering are  $11.7 \times 10^{12}$  and  $12.3 \times 10^{12} \text{ mol yr}^{-1}$ , respectively.

weathering as represented by eq. (6)). It has been argued that increased erosion of continental shelves, estuaries and delta (as sea-level dropped) over the past 40 Myr could have resulted in a decrease in organic carbon burial rate and an increase in the erosion of organic-rich sediments<sup>16</sup>. Such a change in the organic carbon subcycle could prevent a tectonically sustained increase in silicate weathering from completely stripping the atmosphere of  $\text{CO}_2$ . However, somewhat different interpretations and views are provided by France-Lanord and Derry<sup>21</sup>. The sedimentary deposition for Himalayan detritus, the Bengal Fan, has been used to estimate the sinks of  $\text{CO}_2$  from silicate weathering and those from the weathering and burial of organic carbon<sup>21</sup>. It has been demonstrated that the



**Figure 4.** Specific  $\text{CO}_2$  consumption rates for the Himalayan rivers are among the highest compared to global rivers. Rates vary from  $(146 \text{ to } 2250) \times 10^3 \text{ mol CO}_2 \text{ km}^{-2} \text{ yr}^{-1}$ ; with highest consumption rate for the Irrawady and lowest for the Indus (data source – ref. 6).



**Figure 5.** In the global context, specific  $\text{CO}_2$  consumption rate ( $\sim 700 \times 10^3 \text{ mol km}^{-2} \text{ yr}^{-1}$ ) associated with total rock weathering in the Himalaya is about 3 times higher than that for the world average river ( $\sim 250 \times 10^3 \text{ mol km}^{-2} \text{ yr}^{-1}$ ).

lowering of atmospheric CO<sub>2</sub> levels in the Neogene are associated with the enhanced burial of organic carbon in the Bengal Fan. The corresponding CO<sub>2</sub> consumption rates are 2–3 times higher than those derived from the weathering of Himalayan silicates. The flux of organic carbon transported annually to the ocean via global rivers is 0.38 Gt C yr<sup>-1</sup>, nearly 40–50% of which is transported in the particulate form. Thus, the total flux of organic carbon is ~30% higher than the CO<sub>2</sub> flux derived from chemical weathering of rocks<sup>22</sup>. In order to assess the role of continental erosion in the glacial/interglacial carbon cycle, it is therefore important to understand the behaviour and fate of riverine fluxes of organic matter. Based on the empirical models, Ludwig *et al.*<sup>23</sup> have shown that a total of 0.721 Gt C yr<sup>-1</sup> is exported out of the continents to the ocean; of which 0.096 Gt C is derived from carbonate rocks and the remaining 0.625 Gt C results due to the consumption of atmospheric CO<sub>2</sub>. Thus, the variations in the erosion of organic matter and subsequent burial in sediments have the potential to alter atmospheric CO<sub>2</sub> levels over longer time scales.

### Conclusions

On a multi-million year time scale, the continental CO<sub>2</sub> consumption by weathering of silicate minerals is balanced by volcanic degassing, metamorphism and carbonate precipitation, and organic carbon burial. On relatively shorter time scale, atmospheric CO<sub>2</sub> levels are mainly controlled by balance between tectonism and erosion intensity of silicate rocks, as both these driving forces have been varying over the geologic past. This study focuses on the present-day chemical weathering rates in the Himalayan rivers, that could alter the atmospheric CO<sub>2</sub> levels and hence the global climate. The CO<sub>2</sub> consumption rates determined (based on the available data in the literature) for both silicate and carbonate weathering in the six largest rivers of the Himalaya are comparable in magnitude; the total CO<sub>2</sub> consumed by rock weathering is about  $4 \times 10^{12}$  mol yr<sup>-1</sup>. On a global scale, these consumption rates are four times higher than those in the Amazon basin and account for nearly one-sixth of the global CO<sub>2</sub> consumption ( $24 \times 10^{12}$  mol yr<sup>-1</sup>) by continental weathering processes. For comparison, the

specific CO<sub>2</sub> consumption budget ( $\sim 700 \times 10^3$  mol km<sup>-2</sup> yr<sup>-1</sup>) for chemical erosion in the Himalaya is among the highest for global rivers and three times that of average world river ( $\sim 250 \times 10^3$  mol km<sup>-2</sup> yr<sup>-1</sup>). Such higher CO<sub>2</sub> consumption rates have a potential to affect the earth's biogeochemical cycle of C in the present-day scenario of growing anthropogenic CO<sub>2</sub> emission.

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